transition temperature is advantageous, since larger temperature fluctuations can be tolerated without creating a normal zone and, therefore, less heat capacity would be required to dampen temperature fluctuations.

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MODE LOCKING AND ULTRASHORT LASER PULSES BY ANISOTROPIC MOLECULAR LIQUIDS*

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(anisotropic molecular liquids; nitrobenzene; 10^{-11}-sec pulses at 126°C; T/E)

Spectral broadening by the nonlinear dielectric response of anisotropic molecules has been observed and explained by Bloembergen and Lallemand.[1] Any two frequencies, say ω_1 and ω_2, present in the incident optical beam exert a torque on the molecules at a frequency $(\omega_1 - \omega_2)$. The resultant oscillation of the average molecular orientation causes a modulation of the dielectric constant at $\omega_1 - \omega_2$. A third frequency ω_3 experiencing this modulation acquires side-band components at $\omega_3 \pm l(\omega_1 - \omega_2)$ $l = 1, 2, 3, \cdots$.

In this Letter we consider theoretically the case of an anisotropic molecular liquid (henceforth designated as AML) placed within the optical resonator of a multimode laser. In this case the frequencies ω_1 and ω_2 correspond to two longitudinal modes so that the side-band energy at $\omega_3 \pm l(\omega_1 - \omega_2)$ coincides in frequency with already oscillating laser modes. The presence of the AML is thus seen to give rise to power exchange between the equispaced laser modes.

In the following we show that this exchange of energy gives rise, as in the internally modulated laser[2,3] to a phase-locked spectrum characteristic of the ultrashort pulse mode of oscillation, where the pulses are separated by the double transit time $2L/c$ and should approach, in the limit, a duration $\tau \sim (\Delta\nu_{\text{gain}})^{-1}$ where $\Delta\nu_{\text{gain}}$ is the linewidth of the amplifying transition. Such pulses are now obtainable from giant pulse lasers containing a saturable absorber.[4,5]

Our task consists of solving for the complex normal mode amplitudes D_n of an optical resonator containing an AML. These amplitudes are defined as in ref. 3 by the following expansion for the total electric and magnetic fields

$$\bar{E}(\bar{r}, t) = \sum_n i\sqrt{\frac{\omega_n}{2\epsilon}} D_n(t)\, e^{-i\omega_n t}\, \bar{E}_n(\bar{r}) + \text{c.c.}$$

$$\bar{H}(\bar{r}, t) = \sum_n \sqrt{\frac{\omega_n}{2\mu_0}} D_n(t)\, e^{-i\omega_n t}\, \bar{H}_n(\bar{r}) + \text{c.c.} \qquad (1)$$

The summation is over the longitudinal modes whose resonant frequencies are ω_n. $\bar{E}_n(\bar{r})$ and $\bar{H}_n(\bar{r})$ are the vector spatial eigenmodes as defined by Slater.[6] In a linear medium the modes D_n are independent of each other. The presence of an AML spoils this independence and forces a unique phase and amplitude relationship upon the modes.

The mode expansion (1) is substituted into Maxwell's equation. The nonlinear dielectric properties of the medium are accounted for by[1]

$$P^{\omega_3+\omega_1-\omega_2}(\bar{r}, t) = \frac{\epsilon_2}{1 + i(\omega_1 - \omega_2)\tau} \times g(\bar{r}) E_1 E^*_2 E_3\, e^{i(\omega_3+\omega_1-\omega_2)t} + \text{c.c.} \qquad (2)$$

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where ϵ_2 is the optical Kerr constant, τ is the Debye molecular reorientation time, and $g(\bar{r})$ accounts for the partial filling of the resonator volume by the AML. The equations of motion for the modes D_n are obtained by the usual procedure[7] of dot-multiplying Maxwell's curl equations by $\bar{E}_n(\bar{r})$ and $\bar{H}_n(\bar{r})$ and integrating over the resonator volume. The complete analysis is of very considerable difficulty. In order to avoid getting lost in mathematical detail we limit our consideration to all mode pairs ω_1 and ω_2 separated by some multiple, say s, of the intermode frequency ω so that $\omega_1 - \omega_2 = s\omega$. In addition we assume that: (a) a very large number of oscillating modes exists so that spectral end effects are negligible. This assumption can be shown to be equivalent to the neglect of molecular viscous damping; (b) the gain is the same for all the modes and is equal to the loss. If we define the amplitude and phase of D_n through

$$D^*_n(t) = \beta_n(t)\, e^{in\phi(t)} \tag{3}$$

we obtain

$$\beta'(t) + in\phi'(t)\beta = \frac{-3i\omega_n\epsilon_2 f}{2\epsilon^2 V}\left(1 + \frac{1}{1 + s^2\omega^2\tau^2}\right)\mathscr{E}\beta \tag{4}$$

where V is the resonator volume, f is the geometrical filling factor for the AML cell and ϵ is the total stored electromagnetic energy. Since the right side of Eq. (4) is purely imaginary, we obtain the following solution for the normal mode

$$D^*_n(t) = \beta \exp\left\{-i\omega_n t\left[\frac{3\epsilon_2 f\mathscr{E}}{2\epsilon^2 V}\left(1 + \frac{1}{s^2\omega^2\tau^2}\right)\right]\right\}. \tag{5}$$

The AML is thus seen to give rise to a mode-locked spectrum of equal amplitudes and zero phases, so that the optical envelope consists of a train of ultrashort pulses as discussed above. We notice in passing that the factor within the square brackets of Eq. (5) represents frequency pulling which is proportional to the stored energy $\mathscr{E}$.

As a measure of the strength of the mode coupling we define a circulation time T_0 as the exponential time constant for the circulation of the energy in one mode due to its interaction with all the others. Again, if we limit our attention to mode pairs ω_1, ω_2 such that $\omega_1 - \omega_2 = s\omega$, we obtain the partial contribution to T_0^{-1}

$$\left(\frac{1}{T_0}\right)_s = 6\frac{\omega_n\epsilon_2 f\mathscr{E}}{\epsilon^2 V}\left(\frac{s\omega\tau}{1 + s^2\omega^2\tau^2}\right). \tag{6}$$

Since $T_0^{-1} = \Sigma_s\,(T_0^{-1})_s$ we expect from Eq. (6) that the shortest circulation time T_0, hence the strongest mode coupling, obtains when the magnitude of the reorientation time τ satisfies

$$\tau \sim \frac{1}{s_{\max}\omega} = \frac{1}{(\Delta\omega)_{\text{gain}}} \tag{7}$$

where $(\Delta\omega)_{\text{gain}}$ is the gain linewidth of the laser transition.

In an experiment performed in collaboration with J. Comly and Dr. E. Garmire, mode locking and pulses of $\sim 10^{-11}$ sec duration were observed. The setup consisted of a Q-switched ruby laser containing a 5-cm nitrobenzene cell in its optical path. Ultrashort pulses were observed only upon heating the nitrobenzene to $T > 126$°C at which point τ becomes small enough ($\sim 10^{-11}$ sec) to satisfy condition (7). The circulation time T_0 at that point is estimated to be less than 10^{-9} sec so that modes lock together in a time short compared to the duration of a single giant pulse.

A detailed description of the experiment is in preparation.

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